

## REPORT DOCUMENTATION PAGE

AD-A206 410

1b. RESTRICTIVE MARKINGS n.a.		DTIC FILE COPY	
2b. DECLASSIFICATION/DOWNGRADING SCHEDULE n.a.		3. DISTRIBUTION/AVAILABILITY OF REPORT Approved for public release; distribution unlimited.	
4. PERFORMING ORGANIZATION REPORT NUMBER(S) none		5. MONITORING ORGANIZATION REPORT NUMBER(S) ARO 24682.17-CH-UIR	
6a. NAME OF PERFORMING ORGANIZATION Center for the Study of Fast Transient Processes	6b. OFFICE SYMBOL (If applicable) n.a.	7a. NAME OF MONITORING ORGANIZATION U. S. Army Research Office	
6c. ADDRESS (City, State, and ZIP Code) Department of Chemistry University of Southern California Los Angeles, CA 90089-0482		7b. ADDRESS (City, State, and ZIP Code) P. O. Box 12211 Research Triangle Park, NC 27709-2211	
8a. NAME OF FUNDING/SPONSORING ORGANIZATION U. S. Army Research Office	8b. OFFICE SYMBOL (If applicable)	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER DAAL03-86-K-0172	
8c. ADDRESS (City, State, and ZIP Code) P. O. Box 12211 Research Triangle Park, NC 27709-2211		10. SOURCE OF FUNDING NUMBERS PROGRAM ELEMENT NO. PROJECT NO. TASK NO. WORK UNIT ACCESSION NO.	
11. TITLE (Include Security Classification) Center for the Study of Fast Transient Processes - Annual Report			
12. PERSONAL AUTHOR(S) Curt Wittig, Hanna Reisler, Robert Beaudet, and Gerald Segal			
13a. TYPE OF REPORT annual	13b. TIME COVERED FROM 10/87 TO 12/88	14. DATE OF REPORT (Year, Month, Day) December 28, 1988	15. PAGE COUNT 14
16. SUPPLEMENTARY NOTATION The view, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.			
17. COSATI CODES FIELD GROUP SUB-GROUP		18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number) n.a.	
19. ABSTRACT (Continue on reverse if necessary and identify by block number)			
20. DISTRIBUTION/AVAILABILITY OF ABSTRACT <input checked="" type="checkbox"/> UNCLASSIFIED/UNLIMITED <input type="checkbox"/> SAME AS RPT. <input type="checkbox"/> DTIC USERS			
21. ABSTRACT SECURITY CLASSIFICATION Unclassified		22a. NAME OF RESPONSIBLE INDIVIDUAL Dr. Robert Shaw	
22b. TELEPHONE (Include Area Code) (919) 549-0641		22c. OFFICE SYMBOL	

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CRA&I	<input checked="" type="checkbox"/>
DTIC	<input type="checkbox"/>
TAB	<input type="checkbox"/>
Unannounced	<input type="checkbox"/>
Justification	
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## I. Research

### High-energy, collision-induced reactions at surfaces

This work is presently devoted to the study of high-energy collisions between polyatomic molecules and surfaces. A significant advance was made with this experiment during the past year. A year ago, we were at the stage of studying inelastic scattering of small molecules from surfaces under UHV conditions, with state-specific detection of scattered species. We are now able to study collision-induced chemical reactions at several eV (usually  $< 10$ ) incident kinetic energy. Under these conditions, the molecule-surface interaction is impulsive (*i.e.* as near as we can determine, there is little or no 'sticking') and capable of distorting the molecule enough to induce unimolecular decomposition following collision with the surface. This has allowed us to move to larger molecular systems, leaving behind experiments involving vibrational and rotational energy transfer in small species such as NO. Our first experiments on collision-induced dissociation were with  $n\text{-C}_3\text{F}_7\text{NO}$ . The NO fragment was detected state-selectively using 2-photon 2-frequency laser ionization, and it proved possible to obtain high S/N with dissociation probabilities as low as 1%. This work has been submitted for publication and was presented at our ARO Energetic Materials Symposium (August, 1988) and an ACS Meeting (September, 1988). A parallel set of photofragmentation studies with  $\text{C}_3\text{F}_7\text{NO}$  was initiated and showed that many features of the surface-molecule results could be reproduced with photodissociation of the isolated gaseous molecule. This suggests that the energized precursors are similar in the two experiments, and suggests further that the excited molecules may well dissociate via a unimolecular decomposition mechanism. The emphasis with these experiments is to firmly establish the reaction mechanism. If it turns out that the surface is simply vehicular to converting incident kinetic energy into molecular internal energy, then it will be possible to apply the method to a large number of cases in which molecules are excited using this technique in order to bring about reaction.

We were also able to start experiments with 2-methyl-5-vinyl tetrazole (MVT). In its polymeric form, this material is a binder with several desirable properties (*e.g.* desensitization, somewhat energetic), and in monomeric form it has adequate stability and vapor pressure for molecular beam experiments. This was done in collaboration with Dr. Indu Mishra (BRL) who provided the samples and participated in the experiments.

Molecular beams of MVT monomer were prepared by expanding MVT/H<sub>2</sub> mixtures into vacuum, and collisions with MgO(100) surfaces were shown to result in significant internal excitation and *possibly* dissociation. The point here is to examine the different excitation/reaction pathways under conditions of impulsive excitation. The method is applicable to species such as RDX, and, as stated above, provides a clear upper bound to the energy available to the system (*i.e.* the incident kinetic energy). A short internal write-up is available (E. Kolodney, P.S. Powers, H. Reisler, C. Wittig and I.B. Mishra) but these preliminary results are not yet ready for publication. These results were presented at the Energetic Materials Symposium (August, 1988) and an abstract has been submitted and accepted for presentation at the 1989 Detonation Symposium. The work is ongoing and we are optimistic about future prospects. In a recent visit by Dr. Mishra, it was learned that nitrosyl groups *can* be placed selectively in the tetrazoles, thus providing the needed 'chromophore' for establishing dissociation

### Reactions in clusters

During the past year, our studies of bimolecular reactions in clusters changed from systems involving CO<sub>2</sub> (*e.g.* CO<sub>2</sub>-HBr) to those involving nitrogen oxides. Our last paper on the CO<sub>2</sub>-HBr system was written up and will appear soon in Chemical Physics Letters. The first of the nitrogen oxide systems examined was H+N<sub>2</sub>O using N<sub>2</sub>O-HBr complexes, which can react via both NH+NO and OH+N<sub>2</sub> channels. The H+N<sub>2</sub>O reaction is important in combustion, and has been studied both experimentally and theoretically. The low energy channels have been computed *ab initio* and suggest a mechanism (1,3 hydrogen shift to form OH+N<sub>2</sub>) that has far-reaching implications. We have determined several channels under bulk (*i.e.* single-collision, arrested-relaxation) conditions and in complexes, and have measured the [NH]/[OH] ratio. This has been written-up (G. Hoffmann, D. Oh, H. Iams and C. Wittig, Chem. Phys. Lett., (1988) accepted pending minor changes) and a larger paper has been prepared in connection with the Faraday Discussion being held in Bad Honnef, West Germany (12-14 December, 1988). It will be possible to pursue the very fundamental aspects of this work with NSF support and we plan to push the complexes to species such as NO<sub>2</sub>, RNO<sub>2</sub>, etc.

### Spectroscopy

A parallel effort in which high-resolution spectroscopy is used to measure structures of complexes progressed rapidly during the past year. Data acquisition is now quite rapid, and the recording of spectra is routine, except for the fact that the suppliers of

diode lasers (Laser Analytics) is unreliable and it is quite miserable trying to set suitable diodes for our experiments. The analyses of these data are also progressing nicely - the average time to go from a spectrum to a structure is now  $\sim 1$  month. During the past year, we finished an 'initiation rites' project ( $\text{CO}_2\text{-Ar}$ ) and have now recorded spectra of  $\text{CO}_2\text{-HF}$ ,  $\text{CO}_2\text{-HCl}$  and  $\text{CO}_2\text{-HBr}$ . These are being analyzed and the results will be written-up shortly. Spectra have also been recorded for  $\text{N}_2\text{O-Ar}$  and  $\text{N}_2\text{O-N}_2$ . We will do  $\text{N}_2\text{O-HF}$ ,  $\text{N}_2\text{O-HCl}$ , and  $\text{N}_2\text{O-HBr}$  next. The last species have been instrumental in our studies of the  $\text{H+N}_2\text{O}$  reaction. We anticipate continued improvements in the data acquisition and reduction rates. In the near future, we hope to produce complexes consisting of a free radical and a stable molecule.

### Theory

The theory effort in support of these and similar experiments did well last year. About a half dozen projects were completed. These ranged from not-so-relevant starter projects (*e.g.*  $\text{H+ClCN}$ ,  $\text{NCNO}$ ,  $\text{NOCl}$ ) to recent studies of  $\text{H+N}_2\text{O}$  and  $\text{HNO}_3$ .

## II. Equipment

During the past year, several experiments were built/completed for our ARO-sponsored research: (i) A second chamber for the work with complexes was finished this past summer and is now in full operation. (ii) A tunable diode laser spectrometer was built and is in operation except for an insidious RF-pickup problem that we have just recently solved with a screen room. This apparatus was not mentioned in the last section because it did not produce publishable data during the past year. It will be used to study the decomposition of materials such as dimethyl nitramine (*i.e.* the diode laser can find fragments such as NO, NO<sub>2</sub>, OD, etc.). (iii) Our molecule-surface scattering apparatus (also AFOSR sponsored) was modified slightly to accommodate species such as MVT, RDX, and tetranitromethane. (iv) The high-resolution spectroscopy machine has been producing publishable results during the past year and was modified so that spectra can be recorded *very* quickly (*i.e.* a 2 cm<sup>-1</sup> tuning range at 0.0003 cm<sup>-1</sup> resolution can be covered in less than 5 min.). (v) Our photodissociation spectrometer (also NSF sponsored) was moved into new Center space associated with Professor Reisler's tenured appointment. This has provided a data base on the photophysics and photodissociation of nitroso molecules that is used in the surface-molecule collision induced dissociation studies.

### III. Students and postdocs

Student recruiting has been, and remains, a very central part of the Center's activities. More than anything else, it is the enlistment and participation of first-rate graduate students during their formative years that will impact future research in energetic materials. During the past year, a new departmental brochure was developed that explicitly presented the Center as a major entity in Physical Chemistry at USC. In addition, a separate 23 page brochure, *exclusive to the Center*, was developed this past summer, and following examination at the ARO review (Aug. 8) has now been printed. Along with posters having tear-off postcards, they were sent to approximately 250 colleges and universities. Highly-qualified applicants are brought to USC for interviews.

The 1988 Department of Chemistry class of incoming graduate students is the best in its history. This is overwhelmingly true in Physical Chemistry, where 2 NSF predoctoral fellows and one University merit fellow also add to the ARO program. One can never *prove* cause/effect in such a situation. However, Curt Wittig wrote the brochures and is heavily involved in graduate student recruitment, and it is our *reasoned opinion* that the Center's activities and recruitment drive are responsible for a *major change* in the Department of Chemistry's graduate program. There are other side benefits as well. Morale is high, new faculty positions have been made available by the administration, and the increased level of research activity has been instrumental in obtaining new laboratory space (a \$30,000,000 building is envisioned for the early 90's).

**Graduate students and postdoctoral fellows working on ARO-funded projects**

Name	Status	Advisor	Project
Eli Kolodney <sup>(b)</sup>	postdoctoral	Reisler/Wittig	surfaces
Scott Powers	graduate student	Wittig	surfaces
Delroy Baugh	graduate student	Wittig	surfaces
Lori Iwata <sup>(c)</sup>	graduate student	Reisler/Wittig	surfaces
Günter Hoffmann <sup>(a)</sup>	postdoctoral	Wittig	complexes
Daniel Oh	postdoctoral	Wittig	complexes
Yongqin Chen <sup>(a)</sup>	postdoctoral	Wittig	complexes
Helen Iams	graduate student	Wittig	complexes
Steven Sharpe	postdoctoral	Beaudet	spectroscopy
Yongpin Zeng <sup>(d)</sup>	graduate student	Beaudet	spectroscopy
Diane Reifschneider	graduate student	Beaudet	spectroscopy
Raphael Lavi	postdoctoral	Wittig	photodissociation/kinetics
Scott Nickolaisen <sup>(e)</sup>	graduate student	Wittig	kinetics
Amy Ogai <sup>(c)</sup>	graduate student	Reisler	photodissociation
Charles Qian <sup>(a)</sup>	postdoctoral	Reisler	photodissociation
Yi-Yan Bai	postdoctoral	Segal	theory

(a) also supported by NSF

(b) Chaim Weizmann Postdoctoral Fellow, also supported by AFOSR

(c) NSF Predoctoral Fellowship

(d) Teaching Assistant

(e) University Merit Fellowship



#### IV. Communication

During 1988, we had several visitors from Army and other DOD labs and organizations. Some visits were of the information-exchange type and lasted for a day or less [*e.g.* Dr. Richard Paur (ARO) and Dr. Michael Schroeder (BRL)], while others that involved experiments lasted for days or weeks [*e.g.* Dr. Indu Mishra (BRL)]. In the case of the latter, Dr. Mishra made 3 major visits in 1988 in connection with a collaborative experiment involving a material used for binding explosives (2-methyl,5-vinyl tetrazole, MVT). We had developed an 'impact machine' in which molecules are accelerated to high kinetic energies and then collide with a stationary surface; the impact thus generated can initiate different kinds of chemical reactions. These reactions can range from simple bond fission to complex internal rearrangements that involve molecular elimination. The motivation behind the MVT experiments was to (i) see if high-energy impact can lead to dissociation, (ii) see if such dissociation follows the low energy (thermal) channels, (iii) incorporate the binder with energetic materials such as RDX in the gas/surface high energy collision environment. The results, while not yet conclusive, were written as an internal report and have recently been accepted for presentation at the 1989 Detonation Symposium. In the near future, we will examine the collision-induced dissociation of nitroso-substituted tetrazoles, and binder-RDX interactions at the surface, and compare the results to those obtained using thermal decomposition methods.

While it is commonplace for us to have visitors spend a day touring the labs, it is far less common to engage in fruitful collaborative work of the type carried out with Dr. Mishra. Members of the Center also visited Army facilities on several occasions. These visits involved a seminar and informal discussions, usually centered around a specific topic.

C. Wittig visited BRL (February, 1988)  
I.B. Mishra (BRL) visited USC (February, 1988)  
R.A. Beaudet visited BRL (August, 1988)  
H. Reisler visited BRL (July, 1988)  
R.A. Beaudet visited BRL (September, 1988)  
I.B. Mishra (BRL) visited USC (July, 1988)  
R. Paur (ARO) visited USC (September, 1988)  
M. Schroeder (BRL) visited USC (September, 1988)  
I.B. Mishra (BRL) visited USC (December, 1988)

As in 1987, a 2-day research symposium was held following the annual review. This year the conference concentrated on decomposition mechanisms of materials such as RDX, and there were 20 speakers from government, industrial and university laboratories. The conference agenda is given below. The decision to keep the meetings quite topical seems to have paid off. A small group (~ 65) and active audience participation keeps things lively, and there is no sense of competition with the larger meetings.

The Center's newsletter is now distributed to several hundred people every two months at no cost to subscribers. This service provides lists of future conferences and meetings, job opportunities (including postdocs), current mailing addresses (including electronic-mail), and abstracts of publications submitted and/or in press.

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### CHARACTERIZATION AND DIAGNOSTICS OF ENERGETIC MATERIALS

8-10 August, 1988  
University of Southern California

#### Monday, August 8

5:00 - 6:00     Dinner  
7:00 pm        Mixer and social hour

#### Tuesday, August 9

7:00 - 8:00	Breakfast	
8:00 - 8:10	Introductory Comments	
8:10 - 8:45	Carl Melius (Sandia)	Reaction Mechanisms in the Ignition and Combustion of Nitramines.
8:45 - 9:20	Tom Brill (Delaware)	Fast Thermolysis Studies in which Temperature, Mass and Species are Simultaneously Monitored.
9:20 - 9:55	Rich Behrens (Sandia)	Thermal Decomposition of HMX and RDX.
9:55 - 10:15	Break	
10:15 - 10:50	Wing Tsang (NBS)	Important Issues in the Thermal Gas Phase Kinetics of RDX Decomposition.
10:50 - 11:25	Bob Fifer (BRL)	RDX Decomposition and Product Structures Using Tandem Mass Spectroscopy.

11:25 - 12:00	Marilyn Jacox (NBS)	Vibrational and Electronic Spectra of Intermediates in the Decomposition and Combustion of Nitramine Trapped in Solid Argon.
12:00 - 1:00	Lunch	
1:00 - 1:35	Dave Golden (SRI)	Laser Pyrolysis Studies of the Unimolecular Decomposition of Nitramines and Nitroaromatics.
1:35 - 2:10	George Adams (BRL)	Thermochemistry of Boranes and Related Compounds.
2:10 - 2:35	Celeste Rohlffing Michael Koszykowski (Sandia)	Accurate ab initio Predictions of Vibrational Spectra for Energetic Materials Combustion.
2:35 - 3:10	Don Thompson (Oklahoma)	Chemical Dynamics Calculations for Unimolecular Decomposition of Energetic Materials.
3:10 - 3:30	Break	
3:30 - 4:05	Basil Swanson (LANL)	Static-Type Pressure and Shock Studies of Nitrogen Oxides.
4:05 - 4:40	Dick Beyer (BRL)	Studies of Liquid Propellant Drops in Hot, High Pressure Flows.
5:00 - 6:30	Dinner (Mexican-style buffet)	

Wednesday, 10 August

8:00 - 8:35	Lee Harris (ARDEC)	The Low Pressure Combustion of Nitramine Propellant and the Ignition of Low Vulnerability Tank and Artillery Ammunition.
8:35 - 9:00	William Hepler (UCLA)	Numerical Simulation Study of a Hydrazine / Nitrogen Dioxide Diffusion Flame in a Burke-Schumann Burner.
9:00 - 9:35	Tim Edwards (AFAL)	Spectroscopic Measurements of High Pressure Propellant Flames.
9:35 - 10:15	John Vanderhoff (BRL)	Spectral Studies of Propellant Combustion at Moderate Pressures.
10:15 - 10:30	Break	
10:30 - 11:05	Herb Nelson (NRL)	Production and Reactions of Boron Hydrides.
11:05 - 11:40	Dave Chandler (Sandia)	Deuterated Methyl Iodide Photochemistry.
11:40 - 12:10	Curt Wittig (USC)	Photon and Collision-Induced Excitation.
12:00 - 1:00	Lunch	
1:00 - 5:00	Laboratory tours at USC	

## V. List of personnel affiliated with the Center

### *Faculty*

CURT WITTIG  
HANNA REISLER  
ROBERT BEAUDET  
GERALD SEGAL  
SIDNEY BENSON

### *Administrative*

Sarah Novak  
Shirin Mistry<sup>(a)</sup>  
Yuki Yabuta<sup>(a)</sup>

### *Postdoctoral Fellows*

Jeffrey Segall<sup>(d)</sup>  
Julian de Juan<sup>(c)</sup>  
Günter Hoffmann<sup>(b)</sup>  
Eliezer Kolodney<sup>(e)</sup>  
Steven Sharpe  
Brent Koplitz<sup>(c)</sup>  
Diana Schwartz-Lavi<sup>(d)</sup>  
Raphael Lavi  
Daniel Oh  
Yongqin Chen  
Charles Qian<sup>(b)</sup>  
Yi Yan Bai

### *Graduate Students*

Martin Hunter<sup>(h)</sup>  
Helen Iams  
Lori Iwata<sup>(g)</sup>  
David Scott  
Scott Nickolaisen<sup>(f)</sup>  
Amy Ogai<sup>(g)</sup>  
Scott Powers  
Diane Reifschneider  
Carrie Jacques<sup>(h)</sup>  
Yongping Zeng<sup>(h)</sup>  
Wen Ye<sup>(h)</sup>  
Delroy Baugh

(a) Departmental Secretaries

(b) Supported by NSF

(c) Left Summer 1988

(d) Supported by DOE

(e) Supported by AFOSR

(f) University Merit Fellowship

(g) National Science Foundation Predoctoral Fellowship

(h) Teaching Assistant

## VI. Publications (1988)

1.  $\text{H} + \text{ClCN} \rightarrow \text{HCl} + \text{CN}$ : Product excitations and reaction mechanism at  $E(\text{c.m.}) \cong 21.6 \text{ kcal mol}^{-1}$   
J. De Juan, S. Callister, H. Reisler, G.A. Segal and C. Wittig, *J. Chem. Phys.* 89, 1977 (1988).
2. Photoinitiated  $\text{H} + \text{CO}_2 \rightarrow \text{OH} + \text{CO}$ : OH distributions and 3-body interactions in  $\text{CO}_2\text{-H}_2\text{S}$  complexes  
J. Rice, G. Hoffmann and C. Wittig, *J. Chem. Phys.* 88, 2841 (1988).
3. Photoinitiated reactions in weakly-bonded complexes  
C. Wittig, S. Sharpe and R.A. Beaudet, *Acc. Chem. Res.* 21, 341 (1988).
4. Infrared absorption spectroscopy of the  $\text{CO}_2\text{-Ar}$  van der Waals complex  
S.W. Sharpe, R. Sheeks, C. Wittig and R.A. Beaudet, *Chem. Phys. Lett.* 151, 267 (1988).
5. The rotational spectrum, structure, and dipole moment of ethynylphosphine,  $\text{H}_2\text{PC}\equiv\text{CH}$   
E.A. Cohen, G.A. McRae, H. Goldwhite, S. Di Stephano and R.A. Beaudet, *Inorganic Chemistry* 26, 4000 (1987).
6. The molecular structure of boron hydrides and carboranes  
R.A. Beaudet, Molecular Structure and Energetics. Advances in boron and the boranes, J.F. Liebman, A. Greenberg and R.E. Williams, eds., VCH Publishers, NY, chapter 20, pp. 417-490 (1988).
7. Microwave spectrum, structure, and dipole moment of methylaminodiborane,  $\text{B}_2\text{H}_5\text{NHCH}_3$   
S.S. Durso, E.V. O'Gorman, J.R. Chow, A.B. Burg and R.A. Beaudet, *Inorganic Chemistry* 27, 63 (1988).
8. Microwave spectrum, boron skeletal structure, and dipole moment of 5-fluoro, 2,4-dicarbaheptaborane (7),  $\text{C}_2\text{B}_5\text{H}_6\text{F}$   
R.A. Beaudet, *Inorganic Chemistry*, in press (1988).
9. Molecule-surface dissociative scattering of  $n\text{-C}_3\text{F}_7\text{NO}$  from  $\text{MgO}(100)$  at hyperthermal energies: nascent  $\text{NO}(X^2\Pi)$   
E. Kolodney, D. Baugh, P.S. Powers, H. Reisler and C. Wittig, *J. Chem. Phys.*, in press (1988).
10. Singlet and triplet potential energy surfaces of  $\text{NCNO}$   
Y.Y. Bai and G.A. Segal, *Chem. Phys. Lett.*, xxxx (1988).
11. Reactions of hot H atoms with  $\text{N}_2\text{O}$  under single-collision, gas-phase conditions and in photoexcited  $\text{N}_2\text{-HBr}$  complexes:  $\text{OH}(X^2\Pi)+\text{N}_2$ , and  $\text{NH}(X^3\Sigma)+\text{NO}(X^2\Pi)$  channels

G. Hoffmann, D. Oh, H. Iams and C. Wittig, Chem. Phys Lett., accepted pending minor revision (1988).

12. Thermochemistry of methylene phosphine: initial determination of the carbon-phosphorus double bond strength  
J.R. Chow, R.A. Beaudet and H. Goldwhite J. Phys. Chem., in press (1988).
13. State-selective photodissociation dynamics of NOCl: The influence of excited state bending and stretching vibrations  
C.X.W Qian, A. Ogai, L. Iwata and H. Reisler, J. Chem. Phys. 89, 6547 (1988).
14. The electronic spectrum of NOCl: photofragment spectroscopy, vector correlations and *ab initio* calculations  
Y.Y. Bai, A. Ogai, C.X.W. Qian, L. Iwata, G.A. Segal and H. Reisler, J. Chem. Phys., in press (1988).
15. Photoinitiated H + N<sub>2</sub>O reactions: N<sub>2</sub>O-HBr complexes and gas-phase, single-collision, arrested-relaxation conditions  
G. Hoffmann, D. Oh and C. Wittig, Farad. Disc. Chem. Soc., in press (1988).